The Spontaneous Ignition of Under-Expanded Hydrogen Jets Released into Air

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1   Introduction

Hydrogen gas is currently a clean alternative to using gasoline or other fuels for the purpose of generating power. However, a primary concern regarding hydrogen is the safety of storing and handling the fuel. In order for hydrogen to be used as a fuel, it must be compressed to high pressures. This allows a large amount of energy to be available in a small volume. Hydrogen is also highly reactive. The accidental release of this high pressure hydrogen into air can lead to the possibility of spontaneous ignition, without a spark or flame present.

During the release of high pressure hydrogen into air a shock wave is formed. This shock wave travels through the air heating it up to high temperature and pressure. The high temperature induced by the shock wave can trigger ignition in regions behind the shock where the gases have mixed. This phenomenon has been observed experimentally by various groups [1, 2, 3, 4]. Also, during the release process, the gas is expanded and cooled. Therefore, there are two competing mechanisms; one of heat addition due to chemical reactions, and one of cooling due to expansion. In this paper, the two competing mechanisms are studied in order to determine the criteria for ignition to occur during the release of high pressure hydrogen into air.

Current CFD codes [5, 6] for the hydrogen release problem, which take expansion into account, lack the resolution necessary to capture the ignition phenomena that occurs at small scales. In order to study the ignition at the required scales, we introduce an approximate model that reacts a pre-determined, highly reactive, mixture of fuel and air up to the point of ignition while taking realistic expansion rates into account.

2   Governing Equations and Numerical Method

The reaction-diffusion-expansion phenomenon, following a representative Lagrangian particle of fixed mass in the turbulent mixing layer at the head of the jet, is modeled using the *homogeneous mixing ignition* (HMI) method described in [7]. Transport effects are implicitly taken into account by assuming that heat and species diffuse at the same rate. One then has to consider the ignition time history of a fixed mixture fraction in the mixing layer. The ignition of the mixing layer is then assumed to occur at the most reactive mixture fraction of fuel and air. This approximation is well founded on large activation energy asymptotics of ignition-diffusion problems where heat and mass diffuse at the same rate. Also, the approximation assumes that heat release plays a minor role prior to the ignition event. This approach has been shown to be a good approximation for predicting non-premixed hydrogen air ignition problems [7]. The resulting governing equations that are solved numerically for the ignition problem are:

Conservation of mass of the ith specie: $\rho \frac{DY_i}{Dt} = \omega_i$ (1)

Conservation of energy   : $\rho C_p \frac{DT}{Dt} = \frac{Dp}{Dt} - \sum_{i=1}^{N} h_i \omega_i$ (2)

Here, the rate of change of density for specie i per unit time for a fluid particle, $\rho\frac{DY_i}{Dt}$, is controlled by the net rate of production of specie i, ω_i. The rate of change of energy per unit time, per unit volume for a fluid particle, $\rho C_p \frac{DT}{Dt}$, is controlled by the rate of change of pressure per unit time for a fluid particle, $\frac{Dp}{Dt}$, and the energy addition due to the reaction of N species, $\sum_{i=1}^{N} h_i \omega_i$.

The system of coupled equations above is solved explicitly using the Sundials CVODE [8] integrator for the stiff chemistry while the thermodynamic properties are evaluated at each time step using the Cantera [9] libraries for C++. The kinetic mechanism used was developed by [10] and is specifically designed for hydrogen chemistry. Finally the expansion term, $\frac{Dp}{Dt}$, is prescribed as a source term in the energy equation.

3 Initial Conditions and the Shock Tube Problem

The properties of the hot air and cold fuel on each side of the thin mixing layer were found by solving the shock tube problem [11] for the initial conditions of the release. The initial ambient air prior to the release was taken at 1atm and 300K. The initial temperature of the stored hydrogen is also taken at 300K. In order to determine the most reactive mixture fraction in the mixing layer, the initial mixture fraction was varied until the mixture fraction with the smallest ignition delay was found.

4 Expansion Rate

Radulescu and Law [12] have previously determined the scaling parameters for under-expanded non-reactive hydrogen jets. In their non-dimensional analysis, the pressure-time history at the interface between the gasses for different jet conditions was found to be self-similar. The evolution of the mixing layer pressure is shown in Figure 1 in terms of a non-dimensional time τ.

$$\tau = \left(\frac{\rho_{Bo}}{\rho_{Ao}} \left(\frac{2}{\gamma_A+1} \right)^{-1/(\gamma_A-1)} \right)^{1/j} \frac{t a_{Ao}}{R\Lambda} \left(\frac{2}{\gamma_A+1} \right)^{1/2} \tag{3}$$

where a is the speed of sound, γ_A is the ratio of specific heats for the stored hydrogen , t is time, and R is the radius of the hole, (ρ_{Bo} / ρ_{Ao}) is the initial density ratio in the gases and j and Λ are constants whose values are equal to 2 and 1.2 respectively [12].

After an initial transient, there is a unique evolution relation for the pressure of the mixing layer, which is well approximated by

$$\frac{p_i}{p_{Bo}} = 12.2\tau^{-0.68} \tag{4}$$

Furthermore, at the onset of release there is initially a period where the pressure at the interface remains constant. This can clearly be seen for the case where $\frac{p_{Ao}}{p_{Bo}} = 88$ in Figure 1. The reason for this initial period of constant pressure is due to the time required for information regarding expansion at the corner of the hole to reach the location of the particle along the axis of the jet. To account for this delay, the value of τ is calculated for when $\frac{p_i}{p_{Bo}}$ is equal to the initial condition found from the shock tube solution in equation (4).
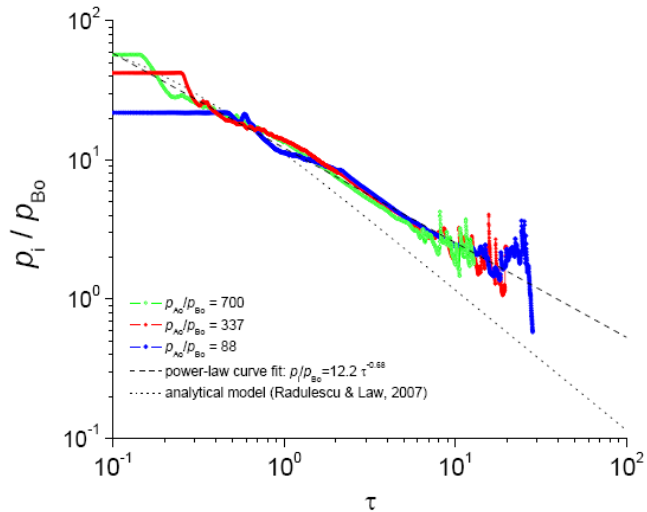

Figure 1. Evolution of the contact surface pressure along the jet axis for round jets obtained numerically for various storage pressure to ambient pressure ratios from Radulescu and Law, 2007 [12].

The simulations are then started at time zero of the release process, incorporating this period of constant pressure.

5   Results, Discussion, and Conclusions

Various jets with pressure ratios ranging from $\frac{p_{Ao}}{p_{Bo}} = 75$ to 800 were simulated with varying hole sizes in order to determine the critical point, or hole size, at which ignition is quenched. The results are shown below in Figure 2 for a case where $\frac{p_{Ao}}{p_{Bo}} = 150$. Results indicate that for large hole sizes, ignition occurs as it would if there were no expansion. As the hole size is reduced, the onset of pressure decay occurs sooner causing a cooling effect in the gas. Despite this cooling effect, however, it is still possible for chemical reactions to occur which can lead to ignition. Ignition is represented in the figure by a sharp increase in temperature. With a further reduction in hole size, a critical point is reached where ignition is not observed. Although there are still chemical reactions that occur on very small scales at this critical point, they do not contribute to any significant large scale changes in the temperature or composition of the gas. Furthermore, by examining Figure 2, the critical point at which the ignition occurs is very sensitive to small changes in the hole size. The smallest change in hole size can lead to full blown ignition, or no ignition at all. This reflects the strong competition between chemical reactions and expansion at this point.
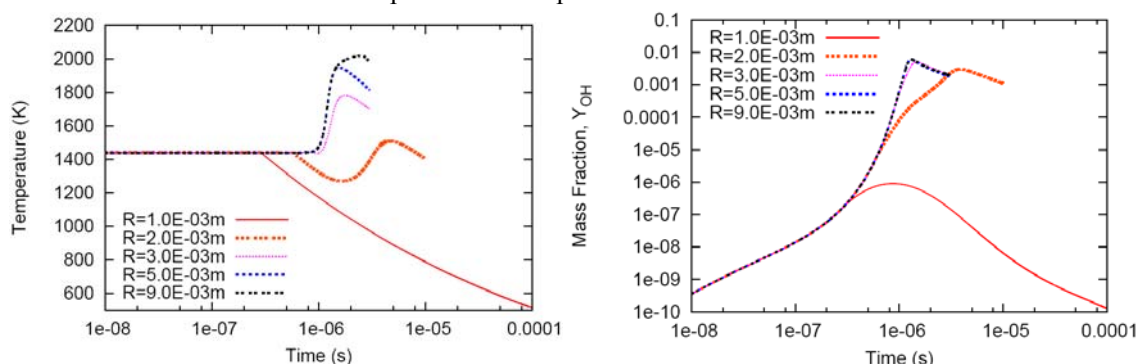

Figure 2. Temperature profiles (left) and OH concentrations (right) for various hole sizes and $\frac{p_{Ao}}{p_{Bo}} = 150$.

The critical hole size for which the onset of ignition occurs for various pressure ratios is shown in Figure 3. It is clear from the figure that higher storage pressures of hydrogen require smaller holes for the gas to escape through in order for ignition to be quenched. This illustrates very well the competition between heating by the lead shock, which is favored for higher pressures, and the expansion cooling, which is stronger in smaller holes, since the cooling rate is faster. It is interesting to note, however, that when the initial pressure of the compressed hydrogen is below 100 atm, the critical hole size becomes very large. Inspection of the initial conditions at the mixing layer and ignition delays indicate that this behavior is due to the proximity to the chain-branching cross-over temperature and pressure.

Critical pressures for ignition for unconfined releases are not available in the literature. Experiments in [2, 3] have however determined the critical ignition pressure for releases through various length pipes. The model is adapted to this geometry by lengthening the residence time of the gas at the shock tube initial state to account for the increase in residence time before the onset of the quenching expansion wave. The approximate model results shown in Figure 4 was found to capture the ignition phenomena within experimental error. However, the large scatter between the experiments conducted suggests that more controlled experiments are required. It should also be noted that the approximate model does not take into account any boundary layer effects within the tubes.
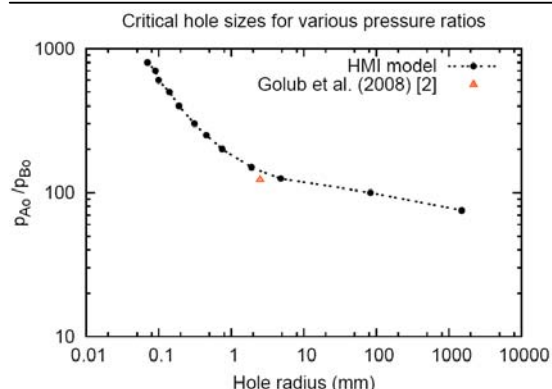
Figure 3. Critical hole sizes for ignition for various pressure ratios (p_{Ao}/p_{Bo}).
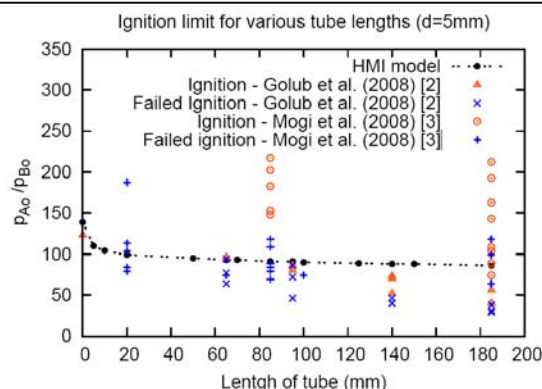
Figure 4. Ignition limit for hydrogen released into air through 5mm diameter tubes of various lengths.

In conclusion, findings in this paper suggest that the ignitability of a high pressure hydrogen jet released into ambient air is governed by two factors; the storage pressure of the hydrogen and the size of the opening through which the hydrogen is released into the air. In particular, the storage pressure controls the strength of the shock that is observed upon release which in turn controls the temperature and pressure of the gases behind the shock. Furthermore, the size of the opening determines the rate at which expansion of the hydrogen gas occurs, or the rate at which the pressure decays at the contact surface between the two gases.

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